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Characterization Report on Fuels for NEAMS Model Validation

Fuel Cycle Research & Development Advanced Fuels Campaign

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ABSTRACT

In order to identify which scattering mechanisms are important in nuclear fuels we performed series of thermal conductivity measurements on selected samples of UO_2 and U_3Si_2. The UO_2 samples were synthesized having different grain sizes and the measurements were performed below room temperature, where grain boundary scattering should dominate. Our detailed studies indicate impact of grain boundary scattering on thermal conductivity in UO_2, especially at low temperatures and in agreement with theoretical modeling. However, before drawing any firm conclusions on this matter more studies are required involving impact of off-stoichiometry and isotope scattering on the thermal transport in this system. By detailed measurements of the electrical resistivity and thermal conductivity we were able to estimate how much heat is carried by electrons and lattice vibrations in U_3Si_2. Our studies indicate that the lattice heat transport is only present at low temperature and is almost entirely governed by electrons above room temperature. These results are important for thermal conductivity calculations where lattice contributions can be neglected at high temperatures.

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1. Impact of Grain Boundary Scattering on Thermal Conductivity of Uranium Dioxide

Nearly 20% of the world's electricity today is generated by nuclear energy from uranium dioxide (UO_2) fuel. The thermal conductivity of UO_2 governs the conversion of heat produced from fission events into electricity and it is an important parameter in reactor design and safety. While nuclear fuel operates at high to very high temperatures, thermal conductivity and other materials properties lack sensitivity to temperature variations and to material variations at reactor temperatures. As a result, both the uncertainties in laboratory measurements at high temperatures and the small differences in properties of different materials inevitably lead to large uncertainties in models and little predictive power. Conversely, properties measured at low to moderate temperatures have more sensitivity, less uncertainty, and have larger differences in properties for different materials. These variations need to be characterized as they will afford the highest predictive capability in modeling and offer best assurances for validation and verification at all temperatures. This is well emphasized in the temperature variation of the thermal conductivity of UO_2. As in other insulating material the grain boundary and defect scattering are dominant at low temperatures, while phonon-phonon umklapp scattering is important at high temperatures.

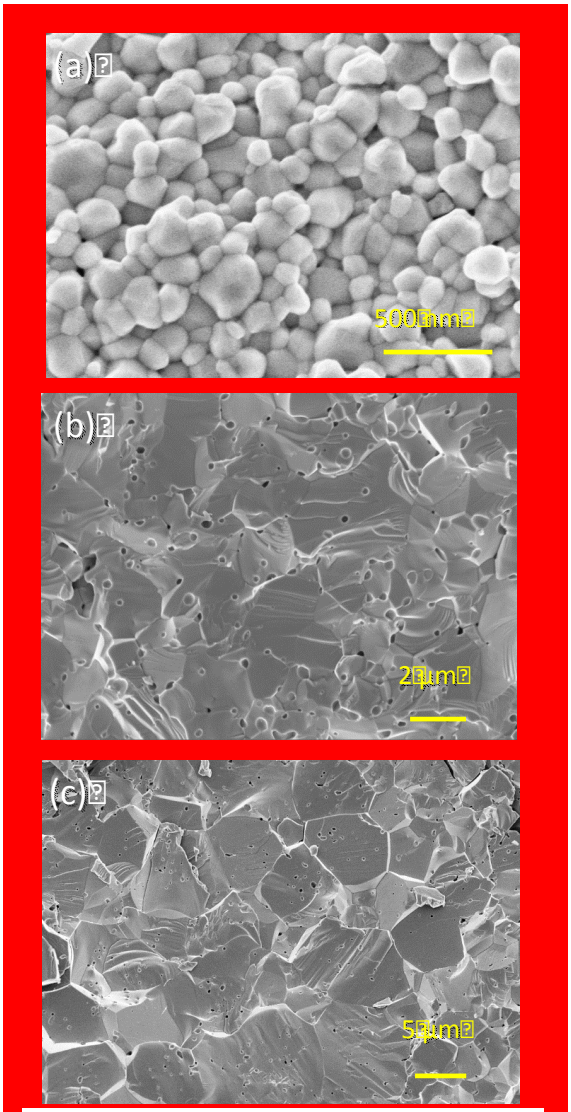

Figure 1 The electron microscope pictures of the UO_2 samples with different grain sizes measured in these studies

In order to better understand the impact of grain boundary scattering on the thermal conductivity of UO_2 fuel we have initiated systematic and extensive investigations of selected samples of uranium dioxide having different grain sizes. The work has been performed in a frame of scientific collaboration with Prof. Jie Lian from Ressenhalser Polytechnic Institute [1]. The uranium dioxide samples have been synthesized using spark plasma sintering method of nano-sized UO_2 powders prepared by high-energy ball milling. Natural uranium has been used for the synthesis. Three samples have been prepared with different sizes of UO_2 grains. Figure 1 shows Scanning Electron Microscope (SEM) pictures of the samples prepared. The average grain sizes estimated were 0.125 µm, 1.8 µm, and 7.2 µm respectively for sample (a), (b), and (c). The so-prepared samples have been measured at INL using direct pulse-power "one heater two thermometers" method implemented in Thermal Transport Option (TTO) on the Quantum Design Physical Properties Measurement System (PPMS). Figure 2 shows one of the UO_2 sample mounted on the TTO setup. The results of the measurements are shown in Figure 3. At room temperature the thermal conductivity is close to 6.8 W/mK for the samples with grain

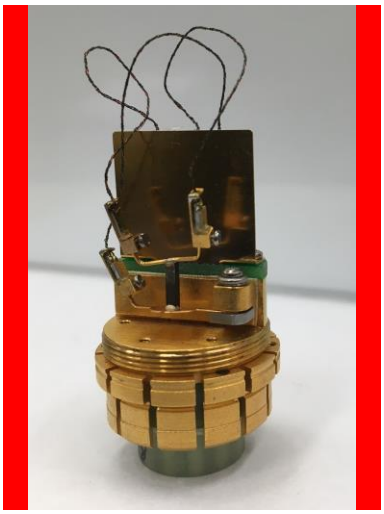

Figure 2 The RPI uranium dioxide sample mounted on QD TTO puck and ready to be measured in PPMS setup

sizes 7.2 and 1.8 μm and it is slightly lower for 0.125 μm sample (6.1 W/mK). These values can be compared with 7.7 W/mK obtained for UO_2 single crystal (ΔT applied along <100> direction). With lowering temperature all samples show the characteristic maximum at around 200 K and sharp minimum at the Neel temperature, T_N = 30.5 K [2]. Below T_N the thermal conductivity increases again and forms a second maximum before going to zero at 0 K as expected for insulating materials. The origin of the low temperature maximum in κ(T) curves of UO_2 is unclear [2] and is a subject of ongoing investigations. At high temperature the polycrystalline samples have similar values and temperature dependence. It is not clear however if the reduced value of κ of the RPI samples, as compared to the single crystal material, can be associated entirely to the gain boundary scattering. The XRD high-angle peak position studies performed on the polycrystalline materials could indicate slight off-stoichiometry of oxygen content. In addition, stronger isotope scattering is expected for the RPI samples since natural uranium has been used for the synthesis compered to dU in the single crystal case. Interestingly, a clear systematic behavior of the κ(T) is observed at very low temperatures, below T_N, where the thermal conductivity is systematically suppressed with the grain size. This temperature range should be less affected by point defects scattering and grain boundaries should dominate as observed in Figure 3. More studies are essential (both experimental and theoretical) to fully understand impact of defect and grain boundary scattering on thermal conductivity of UO_2 and some of them are underway.

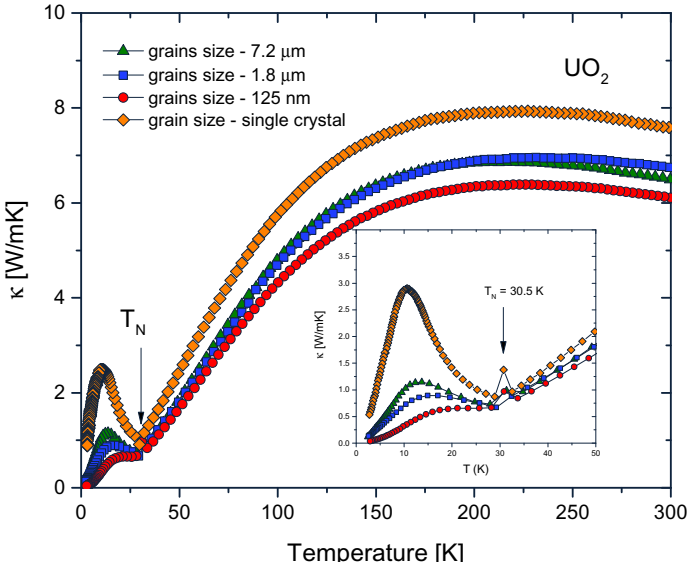

Figure 3 The temperature dependence of the thermal conductivity of RPI samples together with κ(T) of single-crystalline UO_2. Inset: low temperature κ(T). Arrows mark the magnetic phase transition at 30.5 K.

2. Thermal and Transport Properties of Uranium Silicide

Uranium silicide, U_3Si_2, has been considered as a new fuel for the existing Light Water Reactor fleet. This uranium intermetallic has a number of advantageous thermophysical properties, including; high density (12.2 g/cm^3), high thermal conductivity at room temperature (~10 W/m K), and a high melting temperature (1665 ^{o}C) [3]. These properties also support its use as an accident tolerant fuel. Because of its high thermal conductivity, U_3Si_2 can operate at a much lower temperature and experiences lower thermal gradients than UO_2. As a result, it is subject to lower thermal stresses, which should mitigate pellet cracking. In general, the thermal conductivity of intermetallic material is governed by lattice vibrations (phonons) and free electrons.

In U_3Si_2, however, the amount of heat carried by phonons (vs. electrons) was not well accounted. In order to better understand the thermal and transport properties and to estimate how much heat the different heat transport channels carry, we have initiated extensive transport and thermodynamic studies of U_3Si_2 polycrystals. Our preliminary studies are shown in Figure 4. For the thermal conductivity measurements we used the same technique discussed previously for UO_2 samples. As seen from the figure 4 the thermal conductivity of U_3Si_2 has a value of about 8.5 W/mK at room temperature. With lowering temperature the $\kappa(T)$ decreases and forms a broad hump at around 25 K. In order to estimate the electronic contribution to the total thermal conductivity of U_3Si_2 we have measured the electrical resistivity (not shown) and used Wiedemann-Franz relation: $\kappa_{el}(T) = LT/\rho$, where ρ is the electrical resistivity and L is a Lorenz number $L = 2.44 \times 10^{-8}$ $W\Omega K^{-2}$ to calculate $\kappa_{electronic}(T)$. Then, under the assumption of Matthiessen rule concerning different scattering processes we derived the lattice part of total thermal conductivity of U_3Si_2 as $\kappa_{lattice}(T) = \kappa_{total}(T) - \kappa_{electronic}(T)$ [4]. The so obtained electronic and lattice parts of the thermal conductivity of U_3Si_2 are shown in Figure 4 together with the experimental results. As seen, the lattice part is small and can be neglected below room temperature, and electrons, especially above 300 K, carry the vast majority of heat in U_3Si_2. In order to better understand the thermo-physical and thermodynamic properties of this important material we plan to continue our studies in FY2017.

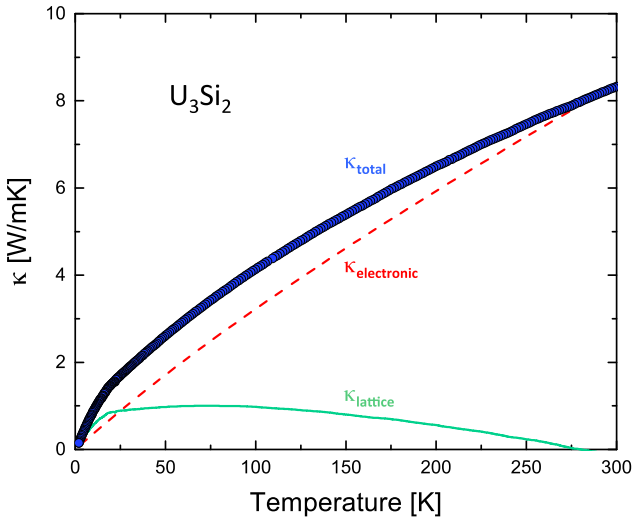

Figure 4 The temperature dependence of the thermal conductivity of U_3Si_2. The dashed and solid lines represent thermal conductivity coming from carriers and lattice vibrations, respectively

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